

Quantum Critical Point in Electron-Doped Cuprates

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Abstract

We analyze doping dependent spectral intensities and Fermi surface maps obtained recently in $\text{Nd}_{2-x}\text{Ce}_x\text{CuO}_{4\pm\delta}$ (NCCO) via high resolution ARPES measurements, and show that the behavior of this electron-doped compound can be understood as the closing of a Mott (pseudo) gap, leading to a quantum critical point just above optimal doping. The doping dependence of the effective Hubbard U adduced by comparing theoretical and experimental spectra is in reasonable accord with various estimates and a simple screening calculation.

Angle-resolved photoemission spectra (ARPES) follow a remarkably different route with doping in the *hole-doped* $\text{La}_{2-x}\text{Sr}_x\text{CuO}_4$ (LSCO) [1] compared to the *electron-doped* NCCO [2]. Starting from a Mott insulator near $x = 0$, LSCO exhibits the appearance of dynamic or static stripes [1], with a concomitant discontinuity in the chemical potential [3]. In sharp contrast, in NCCO, characteristic signatures of stripe order, e.g. the 1/8 anomaly and the NQR wipeout, appear greatly attenuated if not absent [4], and the Fermi level shifts smoothly into the upper Hubbard band [3]. Here we show how the salient features of the most recent high resolution ARPES data in NCCO [2] can be understood in terms of the behavior of a uniformly doped Mott insulator. The doping dependence of the effective Hubbard U parameter adduced from the experimental spectra is in reasonable accord with various estimates, including a computation in which we screen the bare U via interband excitations. A quantum critical point (QCP) where the Mott gap closes is predicted just beyond optimal doping. Finally, we have carried out extensive first-principles simulations of photointensities in order to ascertain that the key spectral features discussed in this article (e.g. the appearance and growth of the $(\pi/2, \pi/2)$ centered hole orbit with doping) are genuine effects of electron correlations beyond the conventional LDA framework and not related to the energy and k-dependencies of the ARPES matrix element [5].

Our analysis of the doping dependence of the Mott gap – perhaps better referred to as a pseudogap [6,7], invokes a mean field solution to the one band [8,9] Hubbard model. The mean field approach provides a good description of not only the pseudogap in the 1D charge density wave (CDW) systems [10], but also of the undoped insulator in the 2D Hubbard problem, including the presence of the ‘remnant Fermi surface’ (rFS) [11] seen in ARPES experiments [12] and Monte Carlo simulations [13]; the spin density wave (SDW) excitations are described by fluctuations about the mean field [14]. The mean field theory correctly predicts a (stripe) phase instability (negative compressibility) associated with hole doping [15]; whereas assuming a finite second-neighbor hopping $t' < 0$, for electron doping the compressibility is positive [16], suggesting that a uniformly doped AFM state should be stable. The preceding considerations argue that the mean field would provide a reasonable model for discussing the doping dependence of the pseudogap. This viewpoint is further supported by mode-coupling calculations [9], which have been applied previously to approximately describe stripes in terms of a CDW in hole-doped cuprates [17,18].

Concerning methodology, we specifically consider the one-band Hubbard model where the Mott gap Δ is well known to arise (in the mean field SDW formulation [14]) from a finite expectation value of the magnetization $m_{\vec{Q}}$ at the wave vector $\vec{Q} = (\pi, \pi)$. The self-consistent gap equation is

$$1 = U \sum_{\vec{k}} \frac{f(E_{\vec{k}}^v) - f(E_{\vec{k}}^c)}{E_{0\vec{k}}}, \quad (1)$$

where f is the Fermi function and

$$E_{\vec{k}}^{c,v} = \frac{1}{2}(\epsilon_{\vec{k}} + \epsilon_{\vec{k}+\vec{q}} \pm E_{0\vec{k}}) \quad (2)$$

Here, the superscript c refers to the upper Hubbard band (UHB) and goes with the $+$ sign on the right side while v goes with the $-$ sign and the lower Hubbard band (LHB) [19]. $E_{0\vec{k}} = \sqrt{(\epsilon_{\vec{k}} - \epsilon_{\vec{k}+\vec{q}})^2 + 4\Delta^2}$, and the independent particle dispersion is: $\epsilon_{\vec{k}} = -2t(c_x + c_y) - 4t'c_x c_y$,

$c_i = \cos k_i a$. We study Eq. 1 self-consistently as a function of electron doping, assuming that the mean-field transition temperature corresponds to the experimentally observed pseudogap. U is thus treated as an effective parameter U_{eff} to fit the experimental data. The Green's function in the antiferromagnetic (AFM) ground state is given by

$$G(\vec{k}, \omega) = \frac{u_{\vec{k}}^2}{\omega - E_{\vec{k}}^c} + \frac{v_{\vec{k}}^2}{\omega - E_{\vec{k}}^v} \quad (3)$$

where

$$u_{\vec{k}}^2 = \frac{1}{2} \left(1 + \frac{\epsilon_{\vec{k}} - \epsilon_{\vec{k}+\vec{Q}}}{2E_{0\vec{k}}} \right), v_{\vec{k}}^2 = \frac{1}{2} \left(1 - \frac{\epsilon_{\vec{k}} - \epsilon_{\vec{k}+\vec{Q}}}{2E_{0\vec{k}}} \right) \quad (4)$$

are the coherence factors.

Using Eqs. 1-4, we discuss spectral density $A(\vec{k}, E)$ (given by the imaginary part of the Green's function) and the related energy dispersions for various doping levels. The FS maps are obtained by taking appropriate cuts through $A(\vec{k}, E)$. Although such theoretical maps do not account for the effects of the ARPES matrix element [5], these maps are relevant nevertheless in gaining a handle on the FS topology expected in the correlated system from ARPES experiments. In any event, we have also carried out first-principles simulations based on the conventional LDA picture where the ARPES matrix element is properly treated and the photoemission process is modeled including full crystal wavefunctions in the presence of the surface – see Refs. [5,20] for details of the methodology.

Fig. 1 displays the doping dependence of the spectral weight in the vicinity of the $(\pi/2, \pi/2)$ and $(\pi, 0)$ points. Fig. 1(a) shows that around $(\pi/2, \pi/2)$, with increasing doping, the spectral weight shifts rapidly towards the E_F as the gap between the LHB and UHB decreases. At $x = 0.15$, the gap is quite small and the LHB and UHB overlap. In contrast, in the momentum region near $(\pi, 0)$, Fig. 1(b), both LHB and UHB are more directly involved. Even at small doping levels ($x = 0.04$), E_F intersects the bottom of the UHB. With increasing electron concentration, the UHB moves to lower energies while the LHB shifts closer to the E_F as the gap decreases. These results are remarkably consistent with the corresponding ARPES data; Fig. 2a of Ref. [2] shows a rapid movement of spectral weight at $(\pi/2, \pi/2)$ from 1.3 eV to around 0.3 eV binding energy close to E_F . This effect, although somewhat less clear, is seen at $(\pi, 0)$ as well (Fig. 2b of Ref. [2]): the weight near E_F grows faster with doping, but the LHB is at a lower binding energy, and is less clearly resolved from the background. This doping dependence suggests that electrons first enter the UHB near $(\pi, 0)$.

The energy dispersions of Fig. 2 are useful not only in understanding the spectra of Fig. 1, but also provide a handle on the FS maps expected in ARPES experiments. The E_F is seen in Fig. 2 to rise smoothly with respect to the UHB with increasing electron doping, whereas in the hole doped cuprates, the E_F gets pinned by the stripes near the mid-gap region over a large doping range. We should keep in mind that various bands do not possess the same spectral weight – this point is emphasized by depicting the coherence factors of Eq. 4 via the width of the bands in Fig. 2. As the Mott gap nearly collapses, the thick lines in Fig. 2(d) essentially present the appearance of the uncorrelated band structure.

Fig. 3 illustrates the evolution of the FS corresponding to the band structure of Fig. 2. For low x , E_F lies near the bottom of the UHB and gives rise to electron pockets centered

at $(\pi, 0)$ and $(0, \pi)$ points in Fig. 3(a). The shadow segments of the FS are clearly evident in Fig. 3(b) where half of the electron pocket gains spectral weight at the expense of the other half due to the decreasing intensity in the magnetic Brillouin Zone (BZ) via the coherence factors; a weak imprint of the magnetic BZ boundary (the diagonal line connecting $(0, \pi)$ and $(\pi, 0)$ points) can also be seen. By $x = 0.15$, the gap shrinks considerably, and the E_F begins intersecting the LHB around $(\pi/2, \pi/2)$. The FS now consists of three sheets: electron-like sheets near $(\pi, 0)$ and $(0, \pi)$ and a hole-like sheet around $(\pi/2, \pi/2)$ separated by a residual gap located at the ‘hot-spots’ [21] along the BZ diagonal. Interestingly, in this doping regime, transport studies find evidence for two band conduction, and a change in the sign of the Hall coefficient [22]; the hole pocket associated with the LHB can explain both these effects.

We stress that the FS evolves following a very different route in the hole-doped case for the $t - t'$ one band Hubbard model [23] (neglecting stripes). At low doping levels, the FS consists of small hole pockets around $(\pi/2, \pi/2)$ points. With increasing hole concentration, these pockets increase in size and merge to yield a large (π, π) centered FS satisfying the volume constraints of the Luttinger theorem.

Figure 4 directly compares the theoretical FS maps against the corresponding experimental results. Here we have included a small second neighbor hopping parameter $t'' = 0.1t$ in the computations in order to account for the slight shift of the center of the hole pocket away from $(\pi/2, \pi/2)$ in the experimental data, even though the maps of Fig. 3 with only $t - t'$ already provide a good overall description (after resolution broadening) of the measurements. The agreement between theory and experiment in Fig. 4 is remarkable: Both sets of maps show the electron pockets at low doping ($x = 0.04$); the beginning of the hole pocket at $x = 0.10$, together with the presence of shadow features around electron pockets and the appearance of intensity around the magnetic BZ; and finally, at $x = 0.15$, the evidence of a well formed hole pocket and a three-sheeted FS which has begun to resemble the shape of a large (π, π) centered hole sheet. Fig. 4 displays discrepancies as well (e.g. the hole pocket related feature appears double peaked in experiments, but not in theory, and other details of spectra around the E_F), but this is not surprising since we are invoking a rather simple one-band Hubbard model and the effects of the ARPES matrix element are missing in these calculations.

The parameters used in the computations of Fig. 4 are as follows. [24] For the half-filled case, the values are identical to those used previously [25]: $t = 0.326\text{eV}$, $t'/t = -0.276$, $U = 6t$. For finite doping x , the only change is that U is assumed to be x -dependent, $U = U_{eff}(x)$, and $t'' = 0.1t$. The actual values of U_{eff} used (solid dots in Fig. 5(a)), are in reasonable accord with various estimates shown in Fig. 5(a) which are: the approximate value from Kanamori (arrow) for a nearly empty band [26], and Monte Carlo results [27,28], for $t' = 0$ (star). Finally, we have carried out a computation of *screened* U using $U_{eff} = U/(1 + \chi P)$, taking P as the charge susceptibility which includes only interband contributions with bare $U = 6.75t$. The U_{eff} so obtained for electrons (solid line) and holes (dashed) is shown [29].

Fig. 5(b) considers the behavior of the staggered magnetization, $m_{\vec{Q}}(x)$, for electrons and holes using the computed values of U_{eff} for *holes* given by dashed line in Fig. 5(a). [30] We see that $m_{\vec{Q}}(x)$ and hence the pseudogap, which is proportional to $m_{\vec{Q}}(x)$, vanishes slightly above optimal doping, yielding a QCP. Notably, superconductivity near an AFM

QCP has been reported in a number of systems [31]. However, the present case is different in that we have a mean-field QCP associated directly with short-range AFM fluctuations and the *closing of the Mott gap*. Since there is no interfering phase separation instability, the bulk Néel transition persists out to comparable, but clearly lower dopings, with $T_N \rightarrow 0$ near $x = 0.13$.

We have carried out extensive first-principles simulations of the ARPES intensities in NCCO within the LDA framework for different photon energies, polarizations and surface terminations, in order to ascertain the extent to which the characteristics of measured FS maps could be confused with the effects of the ARPES matrix element [5] missing in the computations of Figs. 3 and 4. The computed FS maps including full crystal wavefunctions of the initial and final states in the presence of the Nd-CuO₂-Nd-O₂-terminated surface at 16 eV are shown in Fig. 6 for two different polarizations, and are typical. The intensity is seen to undergo large variations as one goes around the (π, π) centered hole orbit, and to nearly vanish along certain high symmetry lines in some cases. Nevertheless, we do not find any situation which resembles the doping dependencies displayed in Figs. 3 and 4. It is clear that strong correlation effects beyond the conventional LDA-based picture are needed to describe the experimental ARPES spectra and that the $t-t'-t''$ Hubbard model captures some of the essential underlying physics.

In conclusion, this study indicates that the electron doped NCCO is an ideal test case for investigating how superconductivity arises near a QCP in doped Mott insulators, untroubled by complications of stripe phases. The doping dependence of U_{eff} adduced in this work has implications in understanding the behavior of the cuprates more generally since the pseudogap in both the electron and the hole doped systems must arise from the same Mott gap at sufficiently low doping.

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FIGURES

FIG. 1. Integrated spectral weight is shown near (a): $(\pi/2, \pi/2)$, and (b): $(\pi, 0)$ points, for four different doping levels, $x = 0.0-0.15$, as marked in (b). Domains in the Brillouin zone over which the spectral weight was integrated are shown in the insets. Fermi energy defines the energy zero in all cases.

FIG. 2. Energy dispersions for various doping levels x . Energy zero defines the Fermi energy as in Fig. 1. Thickness of lines represents the spectral weights of various bands given by the coherence factors of Eq. 4.

FIG. 3. Fermi surfaces corresponding to the band structures of Fig. 2. Maps are obtained by integrating the spectral density function (proportional to $Im[G]$, Eq. 3) over an energy window of 60 meV around E_F ; highs denoted by red and lows by blue. Experimental resolution effects are not included for the illustrative purpose of this figure.

FIG. 4. Upper panels: Theoretical Fermi surface maps including resolution broadening for the $t - t' - t''$ model (see text) for different doping levels x . Lower panels: Corresponding experimental maps after Ref. [2]. Color scheme as in Fig. 3.

FIG. 5. (a): U_{eff} (scaled by nearest neighbor hopping parameter t), and (b): staggered magnetization $m_{\vec{Q}}(x)$, vs doping x for electrons (solid lines) and holes (dashed lines). Filled circles give values used in the computations of Fig. 4 and are representative of the experimental ARPES data in NCCO. Values of U_{eff} given by Kanamori [26] (arrow) and Monte-Carlo studies [27,28] (star) are shown in (a).

FIG. 6. Theoretical FS maps in NCCO obtained via first principles simulations which include the effect of the ARPES matrix element but not of strong correlations for two different polarizations (given by the white arrows) at a photon energy of 16 eV. Color scheme as in Fig. 3.

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